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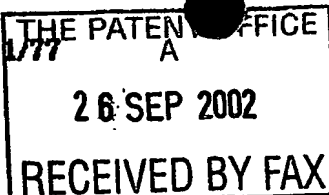
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LRD-GB-1-425

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3. Full name, address and postcode of the or of each applicant (underline all surnames)

K.U. Leuven Research and Development - Groot Begijnhof 59 - 3000 Leuven

Represented by Dr. Ivo Roelants, IPR Officer

Patents ADP number (if you know it)

792460800X
2

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Belgium

4. Title of the invention

Gluten biopolymer articles

5. Name of your agent (if you have one)

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K.U. Leuven R&D

care off:

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Description

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Claim(s)

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Abstract

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DUPLICATE

GLUTEN BIOPOLYMER ARTICLES

5

FIELD OF THE INVENTION

This invention consists of a modified gluten biopolymer for use in industrial applications, such as composites, stabilised foams and molded articles of
10 manufactures.

BACKGROUND OF THE INVENTION

An increase in environmental awareness has given the materials community impetus
15 to develop cost-effective biomaterials with adequate mechanical properties. While research in recent years has led to an improved understanding of the properties of natural fibers, the problem of identifying a cost-effective biopolymer matrix material with suitable properties remains unresolved. This invention describes a method to improve the impact properties of gluten biopolymer, enabling broader usage of gluten
20 in industrial applications.

Experiments designed to improve the impact strength of the gluten material have been previously reported, however these methods require the addition of at least 10-20% (w/w) of some plasticizer, such as glycerol or triethanolamine. In the present work,
25 the fracture toughness of the gluten polymer was improved by a factor of ten or more with the addition of less than 1% (w/w) of a (poly)thiol toughening agent such as a tri-thiol-terminated modifying agent. The toughening agent has another notable attribute, namely, that under proper chemical and environmental conditions, it has the potential to bond chemically with the gluten biopolymer via sulphydryl/disulphide exchange
30 reactions, giving rise to a potentially more stable material (e.g. a stabilised gluten foam). In addition, the process does not require the addition of other solubilizing agents, such as reducing agents or salts, to obtain a homogeneous dispersion. This work also resulted in the development of a gluten biopolymer-modified flax fiber bundle, demonstrating a process to make fully biodegradable composite materials.
35 Qualitative analysis suggests that a reasonably strong interface between the natural

fibers and biopolymer matrix can form spontaneously under the proper conditions, precluding the need to rely on more traditional chemical treatments to promote fiber/matrix adhesion.

5

SUMMARY OF THE INVENTION

The present invention involves a process for improving the impact properties of
10 gluten biopolymer with the inclusion of low levels of a polythiol-terminated hyperbranched molecule to the biopolymer/acid (preferably acetic acid) dispersion, and as modifying agent preferably inclusion of only 1% (w/w) versus gluten or less than 1% (w/w) versus gluten of a polythiol-terminated molecule, preferably a tri-thiol-terminated hyperbranched molecule. The preferred polythiol-terminated
15 hyperbranched molecule is tri-thiol-terminated polyol mercaptoester, such as "TP200 3MP3" (Perstorp Specialty Chemicals AB) (Fig. 1) or an active structural analogue thereof and the preferred acid is acetic acid. The polythiol-terminated hyperbranched molecule above 0.5% (v/v), about 0.5% (v/v), from 0.5% to 0.1 % (v/v) or less than 0.1% (v/v) added to and acetic solution blended with gluten, ideally 10 ml/g.

20 In a further embodiment of present invention a precipitate of modified gluten can be formed by the thiol-terminated hyperbranched molecule, preferably by a tri-thiol-terminated polyol mercaptoester and more preferably by the TP200 3MP3 by increasing the pH of the slurry from 4-4.5 to 6-6.5.

Alternatively a method can be used as described above but wherein an alcohol-based
25 aqueous solution is used, ideally a 50% (v/v) propanol solution in place of the acetic acid solution. Furthermore, the moist modified-gluten chunks (modified by the thiol-terminated hyperbranched molecule, preferably by the thiol-terminated polyol mercaptoester and more preferably by the TP200 3MP3) chunks can be placed in a mold and processed at a variety of different pressures and temperatures (Fig. 2). The
30 pressure can be gradually increased as the water in the gluten network evaporates. The gluten biopolymer can also be used to extrude parts of various geometries (Fig. 3). A further embodiment of present invention is a process to form a cohesive gluten polymer network around fibers by means of the gluten precipitation process. Fibers are covered by gluten powder (ideally 1/1 w/w) and consequently contacted for a

- short period (for instance 30 second) with alkaline water (for instance a pH 11), preferably deionised water with base, preferably NaOH or by contacted said the fibers covered by gluten powder with base water, preferably deionised water with a pH of at least 5 to 6 or higher for instance by dipping said mixture of said fibers and said gluten powder for less than one minute in said water. Figure 4 demonstrates gluten coating fibers, by immersing fibers with gluten powder in an alkaline water bath for less than a minute (Fig. 4). The fibers can be synthetic fibers (e.g. polypropylene fibers or polyethylene fibers) wooden fibers or nonwood fibers (e.g. flax fibers), a combination of wood and nonwood fibers, natural fibers, biodegradable fibers or other fibers comprising cellulose, lignin and/or pentosans. The wood fibers or nonwood fibers can be unmodified natural fibers or can be chemically modified. The gluten biopolymer appears to be bonded to the fibers even after the gluten precipitate is dried (Fig. 5).
- 15 An alternative approach of gluten coating of fibers is to immerse natural wood or non wood fibers in an dilute acid or dilute alcohol gluten solution. The soluble gluten molecules are allowed to interpenetrate the swelling fibers. Optionally polythiol-terminated hyperbranched molecule above 0.5% (v/v), about 0.5% (v/v), from 0.5% to 0.1 % (v/v) or less than 0.1% (v/v) can be added. By increasing the pH or solvent evaporation a strong adhesive bound is formed between the natural fibers and the gluten.

The gluten-coated fibers can be used to reinforce gluten composites by incorporating said gluten coated fibers into heat compression-molded gluten. The gluten-coated fibers can for instance be added to the acid (preferably acetic acid) solution with polythiol-terminated hyperbranched molecule above 0.5% (v/v), about 0.5% (v/v), from 0.5% to 0.1 % (v/v) or less than 0.1% (v/v) and a suspension of gluten, ideally about 10 ml/g to modify said gluten and form a precipitate by increasing the pH, ideally from 4-4.5 to 6-6.5. This fiber/gluten matrix can be compression-molded, for instance for for 10 minutes at 150 °C at a pressure of 200 bars and cooled to form a fiber reinforced gluten polymer article.

Definitions

As used in the specification and the appended claims, the terms "fibers" and "fibrous materials" include both inorganic fibers and organic fibers. In general, fibers can be classified into three categories: wood, nonwood, and nonplant.

Fibers that may be incorporated into gluten matrix preferably include naturally occurring organic fibers, such as cellulosic fibers extracted from hemp, cotton, plant leaves, sisal, abaca, bagasse, wood (both hardwood or softwood, examples of which include southern hardwood and southern pine, respectively), or stems, husks, shells, and fruits or any nonwood fiber as defined hereunder, or inorganic fibers made from glass, graphite, silica, ceramic, or metal materials. Any equivalent fiber which imparts strength and flexibility is also within the scope of the present invention.

The term "nonwood fibers" as used herein is thus to distinguish plant fibers from wood fibers (softwood or hardwood), the fibers can be derived from selected tissues of various mono- or dicotyledonous plants. And are categorized botancially as grass, bast, leaf, or fruit fibers. The nonwood fibers can also be classified by means of production such as sugar cane bagasse, wheat, straw and corn stalks byproducts. They can also be grouped as "fiber plants", plants with high cellulose content that are cultivated primarily for the sake of their fibers such as jute, kenaf, flax, cotton and ramie. An example of nonwood fibers are fibers consisting of the group of Jute, flax, cotton, Hemp, Kenaf, Pina, Abaca, Sisal, Hennequen, Stalk (Rice, Wheat, Barley, Oat, Rye), Cane (Sugar, Bamboo), Grass (Esparto, Sobai), Reed (e.g. *Phragmites communis*), Bast (Seed flax, Kenaf, Jute, Hemp, Ramie), core (Kenaf, Jute), Leaf (Abaca (e.g. Manila), Sisal (e.g. Agave)), Seed hull (e.g. cotton linter).

As other researchers have observed [Gennadios, A., and Weller, C. L. *Food Technol.* 1990, 44, 63-69.-3; Gontard, N., et al J. Food Sci. 1992, 57, 190-195.; Herald, T. J., et al. J. Food Sci. 1995, 60, 1147-1150], the preparation of wheat gluten films necessitates the use of a plasticizer. In the absence of a plasticizer, gluten films are brittle and difficult to handle [Gennadios, A., and Weller, C. L. *Food Technol.* 1990, 44, 63-69.-3; Gontard, N., et al J. Food Sci. 1992, 57, 190-195.; Herald, T. J., et al. J.

Food Sci. 1995, 60, 1147-1150]. A number of plasticizers have been explored in the past, including amines (diethanolamine and triethanolamine) and polyols (anhydrous glycerol, polyethyleneglycols, and polypropyleneglycols). Typical concentrations range from 10g to 60 g/100 g of dry matter [Gennadios, A., and Weller, C. L. *Food Technol.* 1990, 44, 63-69.-3, Roy, S. et al. *J. Food Sci.* 1999, 64, 57-60].

Although there is relatively little in the literature on the use of gluten in industrial applications, wheat gluten films have been studied in significant detail [Gennadios, A., and Weller, C. L. *Food Technol.* 1990, 44, 63-69.-3; Gontard, N., et al. *J. Food Sci.* 1992, 57, 190-195.; Herald, T. J., et al. *J. Food Sci.* 1995, 60, 1147-1150; Roy, S. et al. *J. Food Sci.* 1999, 64, 57-60; Larré, C., et al. *J. Agric. Food Chem.* 2000, 48, 5444-5449]. Films have been cast from gluten protein dispersions in water under different pH conditions or in ethanol [Ali, Y., et al. *Ind. Crops Prod.* 1997, 6, 177-184]. It was also demonstrated that plasticizing agents could be used to improve film flexibility and decrease brittleness [Larré, C., Desserme, C., Barbot, J., and Gueguen, J., "Properties of Deamidated Gluten Films Enzymatically Crosslinked," *J. Agric. Food Chem.* 2000, 48, 5444-5449; Ali, Y., Ghorpade, V. M., and Hanna, M. A., "Properties of Thermally-Treated Wheat Gluten Films," *Ind. Crops Prod.* 1997, 6, 177-184].

20

A separate study focused on understanding the effects of three plasticizers, water, glycerol, and sorbitol, on the glass transition temperature (T_g) of wheat gluten [Pouplin, M., et al. *J. Agric. Food Chem.* 1999, 47, 538]. In summary, water was found to be the most effective plasticizer. At a plasticizer fractional weight content of 0.1, the T_g of wheat gluten decreased from 183°C in its dry state to about 66°C when plasticized with water [Pouplin, M., et al. *J. Agric. Food Chem.* 1999, 47, 538].

25

If we compare the mechanical properties of plain gluten (tensile strength, apparent modulus, and impact strength) with those of several synthetic materials, comprising polypropylene, epoxy, low-density polyethylene (PE), and high-density polyethylene PE (table 1)

30

Polymer	Apparent E-modulus (MPa)	Tensile Strength (MPa)	Charpy Impact Strength (kJ/m ²)
Gluten	3555	43	2.48
Polypropylene	1000-1600	31-37	14
Epoxy	2600-3800	50-79	7.5
Low density PE	100-260	10-12	39
High density PE	410-1240	26-33	68

Table 1. Mechanical properties of various polymers as they compare with gluten matrix material processed at 150 °C/72 bars as measured by the Three-Point-Bend test.

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The apparent E-modulus and tensile strength of wheat gluten seems on par with a number of commonly used synthetic polymers. However, a drawback is the fact that the impact strength of cured gluten is relatively low. The impact strength increased when glycerol was added to the matrix, however this results in the lowering of the E-modulus and tensile strength.

When glutenin chains are synthesized, two cysteine residues on each chain preferentially form interchain disulphide bonds (SS) resulting in linear arrays of polypeptide chains. In the present work, a molecule tri-thiol-terminated polyol mercaptoester, for instance TP200 3MP3 from Perstorp Specialty Chemicals AB, is incorporated into the gluten biopolymer with the intention of crosslinking it with the gluten network, giving rise to a tougher material system. Inherent challenges in processing gluten are attributed to the low solubility of gluten in most solvents, as well as its high melt viscosity.

Illustrative embodiments of the invention

The terminology used herein is for the purpose of describing particular embodiments only, and is not intended to be limit the scope of the present invention.

The thiol molecule, 'TP200 3MP3', was employed in this study due to its flexibility and water compatibility. However, 'TP200 3MP3' can be readily substituted with either 'TP70 3MP3' or 'TMP 3MP3' (Perstorp Speciality Chemicals), which are also available on-site. They differ with respect to degree of ethoxylation; the first carries twenty ethylene oxide units, the second carries seven, and the last does not carry any ethylene oxide units at all.

Gluten powder (150 g) was added slowly (over a period of 1 ½ hrs at room temperature) to 1.5 L 0.05 M acetic acid solution containing 0.1% (v/v) of 'TP200 3MP3'. The mixture was stirred continuously as the gluten powder was added to the solution. The dispersion was put on a shaker in a refrigerator and left overnight. The goal was to obtain a homogenous dispersion of the gluten proteins in the acetic acid, enabling the 'TP200 3MP3' molecule to interact directly with the gluten proteins. The material was recovered by precipitation upon increasing the pH (from 4.5 to 6.5) by addition of dilute NaOH. Afterwards, the gluten/solvent mixture was separated by centrifugation (10,000 g, 20 °C, 15 min) and the modified-gluten precipitate was stored in a refrigerator until further use.

To gain a better understanding of the chemical and physical nature of gluten proteins and how they behave in solution, gluten fractions were also prepared in accordance with a procedure outlined in S Berot et al. [S. Berot, S. et al. Int'l J. Food Science and Technology, (1994), 29, 289-502.] to characterize the various fractions of the gluten biopolymer. Commercial Amylum NV wheat gluten was fractionated to produce gliadin-rich and gluten-rich fractions. Acetic acid solutions (0.01-0.05M) were blended with dry gluten (10 mL/1 g dry gluten) and the slurry was separated by centrifugation (10000 g, 20 °C, 15 min) and spray-dried (compressed air $P = 4$ bars; inlet $T = 130$ °C and outlet $T = 95 - 105$ °C).

In an earlier experiment, a 50% (v/v) propanol/water solution was used in place of the acetic acid solution, and the solvent was evaporated by 'rotavapping' the modified-gluten/solvent mixture at 50 °C. However, subsequent removal of the dried biopolymer from the glass flask was difficult due to the strong adhesion between the gluten network and the glass.

Upon incorporating 0.1 % (v/v) of 'TP200 3MP3' in a 50% (v/v) propanol solution, the fracture toughness of molded gluten was increased from 3 kJ/m² to an average of 36 kJ/m² as measured by the Charpy Impact test (7 Aug. 2002). Individual fracture toughness measurements of four six-day-old TP2003MP3-modified gluten specimens included the following: 47.1 kJ/m², 24.2 kJ/m², 34.2 kJ/m², and 39.2 kJ/m². Specimens were compression-molded at 150 °C/25 bars for 10 min. and subsequently cooled to 20 °C over a period of 5 min. An unmodified control sample yielded a Charpy impact strength of 3.2 kJ/m² (Iso Norm 179 standard analysis).

Legend to the graphics

Fig.1. Schematic of the polyol mercaptoester, TP200 3MP3, $m+n+o = 20$.

Fig. 2. Photograph of an unmodified gluten specimen (left) and TP2003MP3-modified- gluten specimen (right) obtained by the methods described in summary of this invention. These materials were compression-molded for 10 minutes at 150 °C at a pressure of 25 bars. The temperature of the press was subsequently decreased to 20 °C before the mold was removed from the press and the parts demolded.

Fig. 3. Extruded parts of various geometries made from gluten biopolymer.

Fig. 4. A photograph of flax fibers covered with gluten powder, for illustrative purposes only. In the actual experiment, the flax fibers were coated manually by rolling them in the gluten powder. Afterwards, a strainer was used to support the gluten powder-coated fibers as they were immersed in a 45°C alkaline (pH = 11) water bath for 30 seconds.

Fig. 5. A photograph of the gluten biopolymer/flax fiber bundle composite formed after the gluten polymer formed a precipitate around the flax fibers. After two days, the gluten matrix was rigid, as most of the moisture inside the gluten polymer network had evaporated.

Fig. 6. Particle size distributions.

- a. The as-received gluten powder has an average particle diameter of 50 microns.**
- b. The unmodified 0.01 M acetic acid-extracted gluten fraction has an average particle diameter of 20 microns.**

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GLUTEN BIOPOLYMER ARTICLES**Claims**

- 5 1) A toughened gluten polymer matrix, characterized in that it results from precipitating of gluten and modifying the gluten by a polythiol-terminated modifying agent.
- 10 2) The toughened gluten polymer matrix of claim 1, characterised in that the gluten has been precipitated from a watery suspension by an increase in pH.
- 3) The toughened gluten polymer matrix of the claims 1 or 2, characterised in that the polythiol-terminated modifying agent is added to gluten at less 5% (w/w).
- 15 4) The toughened gluten polymer matrix of the claims 1 or 2, characterised in that the polythiol-terminated modifying agent is added to gluten at less 1% (w/w).
- 20 5) The toughened gluten polymer matrix of any of the claims 1 to 4, characterised in that the polythiol-terminated modifying agent is added to gluten in acid dispersion.
- 25 6) The toughened gluten polymer matrix of any of the claims 1 to 4, characterised in that the polythiol-terminated modifying agent is added to a gluten/acetic acid dispersion.
- 7) The toughened gluten polymer matrix of any of the claims 1 to 4, characterised in that the polythiol-terminated modifying agent is added to a dispersion of gluten in alcohol water solution.
- 30 8) The toughened gluten polymer matrix of any of the claims 1 to 4, characterised in that the polythiol-terminated modifying agent is added to a dispersion gluten in propanol water solution.

- 9) The toughened gluten polymer matrix of any of the claims 1 to 4, characterised in that the polythiol-terminated modifying agent is added to the alcohol or acetic acid solution at 0.5% (v/v)
- 5 10) The toughened gluten polymer matrix of any of the claims 1 to 4, characterised in that the polythiol-terminated modifying agent is added to the alcohol or acetic acid solution at 0.5% to 0.1% (v/v).
- 10 11) The toughened gluten polymer matrix of any of the claims 1 to 4, characterised in that an effective amount polythiol-terminated modifying agent is added to the alcohol or acetic acid solution at less than 0.1% (v/v)
- 12) The toughened gluten polymer matrix of any of the claims 1 to 11, characterised in that the dispersion has a gluten load up to 1g/2mL.
- 15 13) The toughened gluten polymer matrix of any of the claims 1 to 12, characterised in that modified gluten is precipitated by increasing to pH from acid to neutral or basic.
- 20 14) The toughened gluten polymer matrix of any of the claims 1 to 13, wherein that the thiol-terminated modifying agent is a polythiol-terminated hyperbranched molecule .
- 25 15) The toughened gluten polymer matrix of any of the claims 1 to 13, wherein that the polythiol-terminated modifying agent is a tri-thiol-terminated hyperbranched molecule
- 30 16) The toughened gluten polymer matrix of any of the claims 1 to 13, wherein that the polythiol-terminated modifying agent is a tri-thiol terminated polyol mercaptoester.
- 17) The toughened gluten polymer matrix of any of the claims 1 to 13, wherein that the thiol-terminated modifying agent is a compound of the group consisting of TP200 3MP3, TP70 3MP3 and TMP 3MP3.

- 18) The toughened gluten polymer matrix of any of the claims 1 to 13, wherein said fiber have been incorporated in said toughened gluten polymer matrix.
- 5 19) The toughened gluten polymer matrix of any of the claims 1 to 13, wherein said fiber, that have been coated by a gluten polymer network precipitated around said fibers, have been incorporated in said toughened gluten polymer matrix
- 10 20) The toughened gluten polymer matrix of any of the claims 1 to 13, wherein said fibers have been incorporated in said toughened gluten polymer matrix, said fibers being precoated by a gluten polymer network precipitated around said fibers by dipping said fibers and said gluten in a water bath with a pH above 5.
- 15 21) The toughened gluten polymer matrix of any of the claims 1 to 13, wherein said fiber have been incorporated in said toughened gluten polymer matrix, said fibers have been interpenetrated by gluten and an adhesive bond has been formed between said fibers and said interpenetrated gluten.
- 20 22) An article of manufacture having the toughened gluten polymer matrix of any of the claims 1 to 17.
- 23) An article of manufacture having toughened gluten polymer matrix of any of the claims 1 to 17 and being fiber-reinforced.
- 25 24) The article of manufacture of claim 23, being enforced by the fibers of any of the claims 19 to 21.
- 25) The article of manufacture of any of the claims 22 to 24, wherein said article is a molded article.

30

Gluten Biopolymer Articles

Abstract

5 This invention consists of a modified gluten biopolymer for use in industrial applications, such as composites and foams. Experiments designed to improve the impact strength of the material have been previously reported, however these methods require the addition of at least 10-20% (w/w) of some plasticizer, such as glycerol or triethanolamine. In the present work, the fracture toughness of the gluten polymer
10 was improved by a factor of ten or more with the addition of preferably less than 1% (w/w) of a tri-thiol-terminated modifying agent. The toughening agent has another notable attribute, namely, that under proper chemical and environmental conditions, it has the potential to bond chemically with the gluten biopolymer via sulphydryl/disulphide exchange reactions, giving rise to a potentially more stable
15 material. In addition, the process does not require the addition of other solubilizing agents, such as reducing agents or salts, to obtain a homogeneous dispersion. This work also resulted in the development of a gluten biopolymer-modified flax fiber bundle, demonstrating the potential to process fully biodegradable composite materials. Qualitative analysis suggests that a reasonably strong interface between the
20 natural fibers and biopolymer matrix can form spontaneously under the proper conditions, precluding the need to rely on more traditional chemical treatments to promote fiber/matrix adhesion.

FIGURES

Fig.1.

5

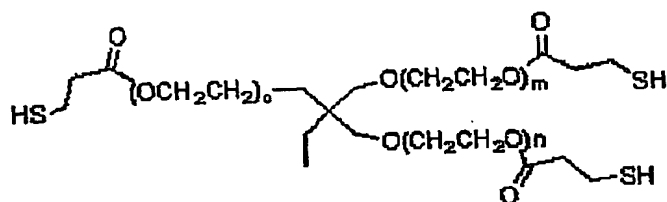


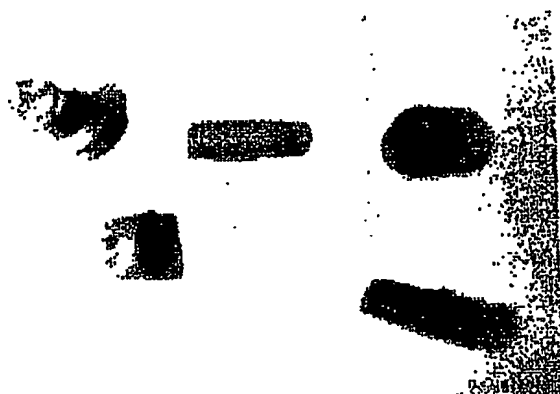
Fig. 2.

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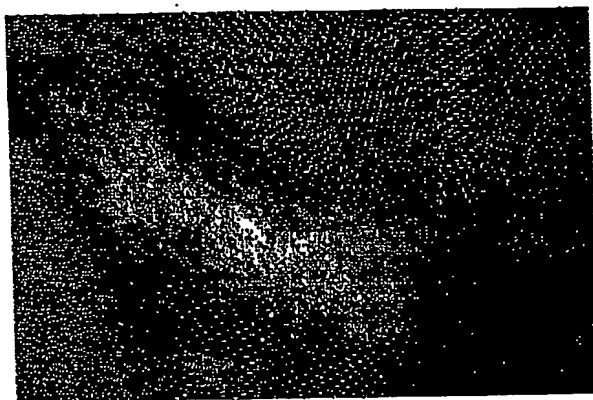
Fig. 3.



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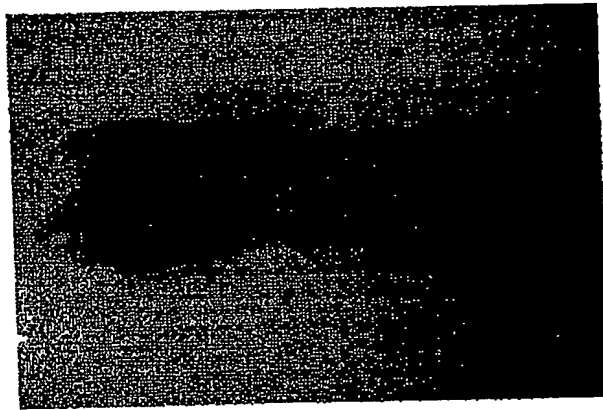
Fig. 4

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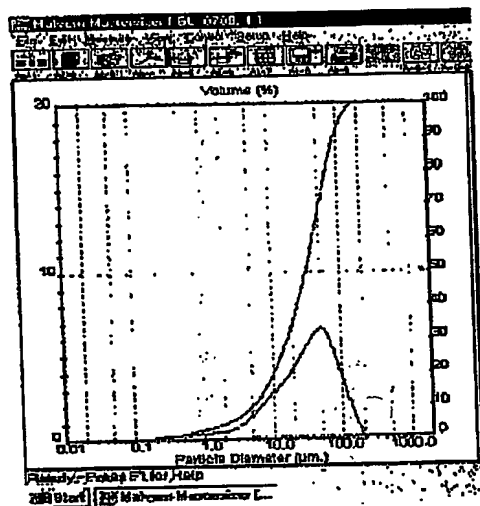
Fig. 5.



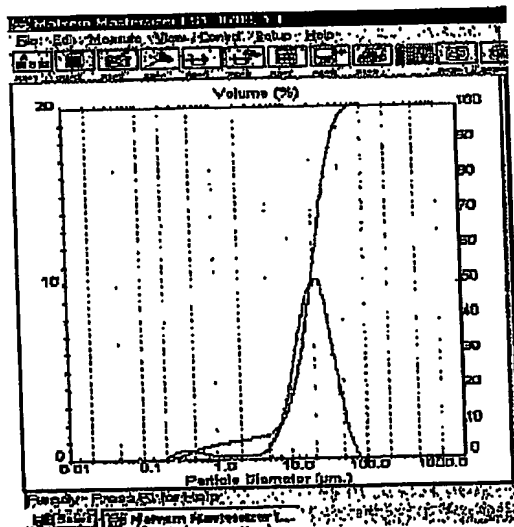
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Fig. 6. A

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10 Fig. 6 B



PCT Application

BE0300163

